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Characteristics of organic compounds in aerosol particulate matter from Dhahran city, Saudi Arabia

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Abstract Organic chemical pollutants in atmospheric particulate matter (PM) have a potential toxicity hazard resulting in human responses that vary from no discernible effect to premature death. The formation and sources of PM also affect air quality of metropolitan areas as well as climate change. The new developments and industrial activities in the Middle East, especially Saudi Arabia, are expected to contribute to the natural, regional, and anthropogenic input sources of organic matter (OM). Here we report the occurrence, concentrations and sources of organic tracers, including *n*-alkanes, polycyclic aromatic hydrocarbons (PAHs), plasticizers, and petroleum biomarkers, in ambient atmospheric PM from the city of Dhahran, Saudi Arabia. The major compounds were unresolved complex mixtures (UCM) of branched and cyclic hydrocarbons (489 ± 296 ng m^{-3}), plasticizers (131 ± 119 ng m^{-3} for phenyl phosphates, 87 ± 42 ng m^{-3} for phthalates), *n*-alkanes (73 ± 53 ng m^{-3}), hopane biomarkers (11 ± 8 ng m^{-3}), *n*-alkanones (6.7 ± 6.3 ng m^{-3}), PAHs (2.0 ± 2.1 ng m^{-3}), *n*-alkanols (1.2 ± 1.2 ng m^{-3}), sterane biomarkers (0.4 ± 0.3 ng m^{-3}), and sterols (0.5 ± 0.4 ng m^{-3}). Obviously, UCM and plasticizers were the major components (56 ± 9% and 26 ± 10% of the total extracts, respectively) in the PM of Dhahran, which might have adverse public health effects. The major sources of this OM are emissions from industrial factories north of the city, plastics and biomass burning, and petroleum product combustion (traffic/refining).

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1. Introduction

A wide-range of contaminants have been identified in atmospheric particulate matter (PM) (Arimoto et al., 2004; Bardouki et al., 2003; Bi et al., 2008; Capes et al., 2008; Didyk et al., 2000; Johansen et al., 2000; Kolb and Worsnop, 2012; Pio et al., 2008; Schauer et al., 1996; Simoneit, 1977, 1999; Simoneit et al., 2004; VanCuren, 2003). They include inorganic compounds such as minerals with associated major and trace elements (Arimoto et al., 1996; Bardouki et al., 2003; Chester et al., 1991; Johansen et al., 2000; Khemani et al., 1985); organic compounds from fossil fuel use and combustion, synthetic organic compounds such as pesticides, chlorinated hydrocarbons, lubricants, and natural product compounds from biota (Alfarra et al., 2007; Medeiros and Simoneit, 2008; Rogge et al., 1996; Schauer et al., 1996; Simoneit, 1984, 1989, 1999; Williams et al., 2006); and radioactive materials (Butler, 2011; Wheeler, 1988).

Atmospheric dust and fine aerosol components introduce a wide variety of materials with different chemical compositions to air (Chester et al., 1991; Didyk et al., 2000; Khemani et al., 1985; Simoneit, 1977, 1978; Zhou et al., 2008, 2009). The Arabian Peninsula is one of the largest sources of aerosol particulate matter (PM) in the world, because it has natural dust advection and different anthropogenic inputs (e.g. traffic and industry emissions, biomass and agricultural waste disposal). Rushdi et al. (2010, 2013) reported the detailed organic molecular composition of fine dust from the region (e.g. Saudi Arabia and Kuwait). They identified fossil fuel combustion and plasticizers as significant fractions of the organic compounds in fine dust particles in the region.

Organic components in aerosol PM also affect human health, because of their carcinogenic effects (Brown et al., 1996; Hannigan et al., 1998; Kawanaka et al., 2004). Due to the high variability in composition, aerosol organic matter needs to be defined to identify its various sources. Air quality of Saudi Arabia is important and under increasing deterioration due to increasing human activities (Rushdi et al., 2010, 2013). Information and data are still lacking regarding the characteristics, levels and sources of pollutants in aerosol PM of the major cities in Saudi Arabia.

The eastern part of Saudi Arabia is a major industrial area with limited information on the level of pollution. Therefore, the purpose of this work is to investigate the characteristics, compositions and levels of organic compounds in atmospheric PM from the city of Dhahran, Saudi Arabia. This is an initial investigation on the concentrations of different compound classes including aliphatic hydrocarbons, synthetic organic compounds, PAHs and unresolved complex mixture (UCM) in aerosols of the eastern province of Saudi Arabia. The possible major emission source categories of the organic tracers in aerosol PM are also reported.

2. Study area

Dhahran, which was selected for this study, is located about 98 km south of Al-Jubail, an industrial city (Fig. 1). Al-Jubail is considered as the largest industrial complex in the Middle East, and an important regional commercial and industrial locale for different manufacturers. Besides a steel mill, it has an oil refinery and one of the world's largest desalination plants. The city of Al-Jubail also generates electric power. Petrochemical products such as plastics and fertilizers are also manufactured there. Other industries were also established to manufacture consumer goods and provide support to the activities of the oil- and gas-based companies. Al-Jubail is also one of the most important ports of Saudi Arabia.

The climate of the eastern part of Saudi Arabia is tropically hot and relatively humid. Usually, the highest average temperature (~43 °C) is in July and the coldest average temperature (~10 °C) is in January. The highest humidity (~73%) is generally observed during January and lowest in August.

3. Experimental procedures

3.1. Sampling

A standard high volume sampler was used for the collection of the aerosol PM samples on quartz fiber filters (QMA, 20.3 × 25.4 cm). The sampler was installed at about 10 m above the ground level on the roof of the King Fahd University for Petroleum and Minerals (KFUPM). Before sampling, the filters were heated to 600 °C to lower their background levels and then placed in cleaned pre-extracted aluminum containers. Weighed filters were put in the sampler and PM aerosol samples were collected at a flow rate of 1.2 m^3 min^{-1} for about 24 h. A total of 7 PM samples were obtained during the months of June to September of 2011 for analysis of organic compounds. The filters were wrapped with aluminum foil, placed in a polyethylene container and brought to the laboratory after sampling. The filters were transferred into desiccators, then weighed, and stored at −20 °C until analysis. A sample blank was also acquired for analysis.

3.2. Extraction

Filters were solvent-extracted with a mixture of dichloromethane (DCM) and methanol (MeOH) (2:1 v:v) using a stainless steel Accelerated Solvent Extractor (ASE) cell (Dionex, Sunnyvale, CA, USA). Prior to use each ASE cell was pre-cleaned by rinsing with organic solvents and baked over-night. Each ASE cell with a filter was filled with anhydrous sodium sulfate and capped. Extraction was affected with the solvent mixture at 100 °C, 1500 psi, 2 cycles of 5 min and 150% flush volume. The extract was concentrated to 0.5 mL in a TurboVap II (Zymark, Hopkinton, MA) with nitrogen. Aliquots of typically 50 μL of these total extracts were treated with N,O-bis-(trimethylsilyl)trifluoroacetamide (BSTFA) containing 1% trimethylchlorosilane and pyridine in a vial with a Teflon-lined cap for 3 h at 70 °C to convert polar compounds to trimethylsilyl derivatives prior to gas chromatography–mass spectrometry (GC–MS) analysis.

3.3. Instrumental analysis

Instrumental analysis by GC–MS was carried out with an Agilent 6890 gas chromatograph coupled to a 5975C Mass Selective Detector, using a DB-5MS (Agilent) fused silica capillary column (30 m × 0.25 mm i.d., 0.25 μm film thickness), and helium as carrier gas. The GC was temperature programmed from 65 °C (2 min initial hold) to 310 °C at 6 °C min^{-1} (isothermal for 20 min final time) and the MS was operated in

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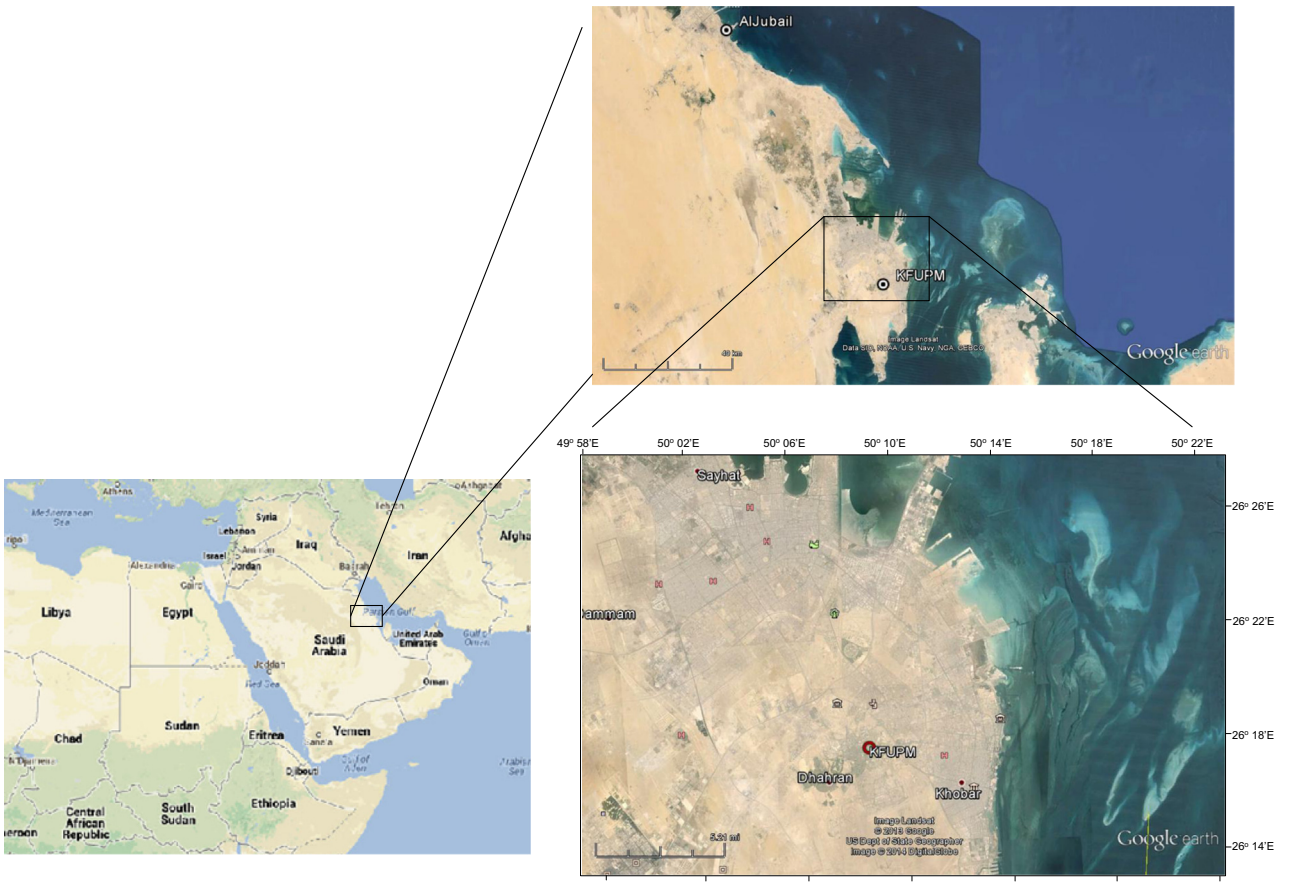

Figure 1 Location map of the sampling site in Dhahran (sample collection) and the industrial city of Al-Jubail.

the electron impact mode at 70 eV ion source energy. Mass spectrometric data were acquired and processed using the GC–MS ChemStation data system.

3.4. Identification and quantification

The identification of *n*-alkanes, *n*-alkanols, methyl *n*-alkanoates, *n*-alkanones, hopanes, steranes, phenyl phosphates, and phthalates is based primarily on their MS key ion pattern (i.e., fragmentograms *m*/*z* 85, 103, 87, 58, 191, 217/218, 326/340/354/368 and 149, respectively) and gas chromatographic retention times. Retention times were compared with those of external standards. Quantification was performed from the total ion current (TIC) GC profiles using the external standard method with authentic compounds of each homologous series (Rushdi et al., 2006, 2010). Average response factors were calculated for each compound. All quantifications were based on the peak areas of the compounds derived from the TIC trace.

3.5. Quality control

Emphasis has been placed on the technical accuracy of the research, including the sample collection, analytical chemistry, and biomarker approach and geochemical application throughout this study. The solvents and the reagents used in sample extractions and the analytical workup procedure have been tested for possible contaminants. Qualitative and quantitative assessment of background contamination introduced by laboratory procedures has been examined by procedural blanks. Also, blank extracts were performed through the entire procedures after batches of three samples.

4. Results and discussion

The mass distribution and the concentrations of the organic compounds identified in the aerosol PM from the Dhahran are given in Table 1. The mean mass concentrations of total PM were 400 ± 383 μg m^{-3}. The major compound groups for the urban samples were unresolved complex mixture (UCM) of branched and cyclic hydrocarbons, synthetic organic compounds (mainly plasticizers), *n*-alkanes, *n*-alkanones, methyl *n*-alkanoates, and polycyclic aromatic hydrocarbons (PAHs). These compound groups are described in detail below.

4.1. Hydrocarbons

Both UCM and *n*-alkanes (e.g. Figs. 2a and 3a) were found at high levels and comprised from 45% to 72% (mean = 65 ± 12%) of the total extractable organic matter (EOM) (Table 1), indicating inputs from petroleum derived lubricants and related petroleum product utilization by traffic (Simoneit, 1984, 1985). Carbon preference index (CPI) values of *n*-alkanes can be used as a diagnostic surrogate to identify their sources

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Table 1 Concentrations (ng m^{-3}) and parameters of the major compounds in aerosol particulate matter from the city of Dhahran for June/July and August/September 2011.

Compound	Composition	M.W.	Sampling date							Mean	SD
			23/6	3/7	6/7	20/7	17/8	8/9	14/9		
n-Alkanes											
Tetradecane	$C_{14}H_{30}$	198	0.3	0.0	0.0	0.0	0.0	0.0	0.0		
Pentadecane	$C_{15}H_{32}$	212	0.3	0.0	0.0	0.0	0.0	0.0	0.0		
Hexadecane	$C_{16}H_{34}$	226	0.9	0.0	0.0	0.0	0.3	0.0	0.0		
Heptadecane	$C_{17}H_{36}$	240	2.4	0.4	0.0	0.6	2.3	0.0	0.8		
Octadecane	$C_{18}H_{38}$	256	2.9	0.5	0.2	0.7	2.3	1.1	2.0		
Nonadecane	$C_{19}H_{40}$	268	4.5	0.2	0.2	1.2	3.0	0.3	1.2		
Eicosane	$C_{20}H_{42}$	282	7.0	0.7	1.3	2.1	5.8	0.6	1.0		
Heneicosane	$C_{21}H_{44}$	296	9.6	0.4	1.5	2.3	6.0	0.8	2.9		
Docosane	$C_{22}H_{46}$	310	11.2	0.9	2.4	4.1	8.6	1.2	4.4		
Tricosane	$C_{23}H_{48}$	324	13.5	1.2	3.6	4.6	10.4	1.8	5.8		
Tetracosane	$C_{24}H_{50}$	338	16.8	1.8	5.4	6.4	12.9	2.4	6.9		
Pentacosane	$C_{25}H_{52}$	352	15.9	1.6	3.3	6.2	13.1	2.5	5.0		
Hexacosane	$C_{26}H_{54}$	366	14.6	1.3	5.7	6.3	11.8	2.6	4.0		
Heptacosane	$C_{27}H_{56}$	380	13.3	1.5	4.7	8.3	14.8	3.5	3.6		
Octacosane	$C_{28}H_{58}$	394	8.1	0.9	3.6	4.6	7.7	2.2	1.8		
Nonacosane	$C_{29}H_{60}$	408	13.6	2.5	3.7	13.6	18.2	5.5	4.4		
Triacontane	$C_{30}H_{62}$	422	4.0	0.5	2.0	2.6	3.9	1.6	1.0		
Hentriacontane	$C_{31}H_{64}$	436	9.6	1.8	2.3	10.3	11.4	4.1	3.1		
Dotriacontane	$C_{32}H_{66}$	450	1.7	0.2	0.7	1.2	1.7	0.9	0.5		
Tritriacontane	$C_{33}H_{68}$	464	2.1	0.3	0.5	1.9	2.2	1.2	0.8		
Tetratriacontane	$C_{34}H_{70}$	478	0.5	0.0	0.2	0.4	0.5	0.4	0.2		
Pentatriacontane	$C_{35}H_{72}$	492	0.3	0.0	0.1	0.3	0.4	0.3	0.1		
Hexatriacontane	$C_{36}H_{74}$	506	0.0	0.0	0.0	0.0	0.0	0.1	0.0		
Heptatriacontane	$C_{37}H_{76}$	520	0.0	0.0	0.0	0.0	0.0	0.1	0.0		
Total			153.3	16.7	41.4	77.8	137.1	33.4	49.5	73.0	53.0
% of total EOM			10.9	5.9	3.2	15.3	12.6	9.6	5.2	9.0	4.4
CPI (o/e)[a]			1.25	1.46	0.93	1.73	1.48	1.55	1.26	1.38	0.26
CPI (o/e) (24–35)			1.20	1.62	0.83	1.89	1.56	1.71	1.18	1.43	0.37
Pristane	$C_{19}H_{40}$	268	0.26	0.01	0.01	0.24	0.65	0.34	0.08	0.23	0.23
Phytane	$C_{20}H_{42}$	282	0.78	0.07	0.06	0.21	0.56	0.28	0.54	0.36	0.28
Pr/Ph			0.33	0.11	0.20	1.17	1.17	1.20	0.16	0.62	0.53
Plant Wax			20.1	4.2	3.9	23.80	30.0	9.2	7.0	14.04	10.48
% of total EOM			1.4	1.5	0.3	4.7	2.8	2.7	0.7	2.01	1.49
Hopane Biomarkers											
Trisnorneohopane	$C_{27}H_{46}$	370	0.90	0.00	1.17	0.54	0.76	0.13	0.21		
17α(H)-Trisnorhopane	$C_{27}H_{46}$	370	0.67	0.01	0.32	0.45	0.55	0.14	0.16		
17αH),21β(H)-Norhopane	$C_{29}H_{50}$	398	3.46	0.03	5.72	3.63	3.41	0.85	0.80		
17α(H),21β(H)-Hopane	$C_{30}H_{52}$	412	3.34	0.04	5.10	4.35	3.05	1.09	0.69		
17α(H),21β(H)-22S-Homohopane	$C_{31}H_{54}$	426	1.97	0.03	2.65	2.69	1.91	0.79	0.42		
17α(H),21b(H)-22R-Homohopane	$C_{31}H_{54}$	426	1.47	0.02	2.00	2.11	1.48	0.57	0.30		
Gammacerane	$C_{30}H_{52}$	412	0.24	0.01	0.31	0.31	0.12	0.03	0.02		
17α(H),21β(H)-22S-Bishomohopane	$C_{32}H_{56}$	440	1.02	0.02	1.27	1.39	0.98	0.23	0.23		
17α(H),21β(H)-22R-Bishomohopane	$C_{32}H_{56}$	440	0.71	0.01	0.86	0.90	0.62	0.13	0.14		
17α(H),21β(H)-22S-Trishomohopane	$C_{33}H_{58}$	454	0.59	0.01	0.69	0.81	0.52	0.10	0.12		
17α(H),21β(H)-22R-Trishomohopane	$C_{33}H_{58}$	454	0.37	0.01	0.48	0.45	0.34	0.07	0.08		
17α(H),21β(H)-22S-Tetrakishomohopane	$C_{34}H_{60}$	468	0.28	0.01	0.34	0.40	0.26	0.08	0.06		
17α(H),21β(H)-22R-Tetrakishomohopane	$C_{34}H_{60}$	468	0.20	0.01	0.21	0.27	0.17	0.04	0.04		
17α(H),21β(H)-22S-Pentakishomohopane	$C_{35}H_{62}$	482	0.18	0.01	0.24	0.31	0.22	0.00	0.07		
17α(H),21β(H)-22R-Pentakishomohopane	$C_{35}H_{62}$	482	0.12	0.00	0.14	0.13	0.14	0.00	0.04		
Total			15.5	0.2	21.5	18.7	14.5	4.3	3.4	11.17	8.39
C_{31} S/(R + S)			0.57	0.60	0.57	0.56	0.56	0.58	0.59	0.58	0.01
% of total EOM			1.1	0.1	1.7	3.7	1.3	1.2	0.4	1.35	1.17
C_{32} S/(R + S)			0.59	0.63	0.59	0.61	0.61	0.64	0.63	0.61	0.02
Steranes											
13β,17α-20S-Cholestane (Diasterane)	$C_{27}H_{48}$	372	0.01		0.01	0.04	0.05	0.02	0.04		
13β,17α -20R-Cholestane (Diasterane)	$C_{27}H_{48}$	372	0.01		0.02	0.02	0.03	0.01	0.02		
13α,17β-20S-Cholestane (Diasterane)	$C_{27}H_{48}$	372	0.01		0.02	0.01	0.00	0.00	0.00		

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Table 1 (*continued*)

Compound	Composition	M.W.	Sampling date							Mean	SD
			23/6	3/7	6/7	20/7	17/8	8/9	14/9		
13α,17β -20R-Cholestane (Diasterane)	$C_{27}H_{48}$	372	0.02		0.05	0.02	0.00	0.00	0.00		
14α,17β-20S-Cholestane	$C_{27}H_{48}$	372	0.02		0.05	0.03	0.04	0.03	0.01		
14β,17β-20R-Cholestane	$C_{27}H_{48}$	372	0.05		0.03	0.05	0.08	0.02	0.03		
14β,17β -20S-Cholestane	$C_{27}H_{48}$	372	0.04		0.03	0.03	0.04	0.01	0.01		
14α,17β-20R-Cholestane	$C_{27}H_{48}$	372	0.03		0.00	0.04	0.05	0.02	0.02		
14α,17β -20S-Ergostane	$C_{28}H_{50}$	386	0.01		0.02	0.00	0.01	0.01	0.01		
14β,17β -20R-Ergostane	$C_{28}H_{50}$	386	0.01		0.03	0.02	0.02	0.01	0.01		
14β,17β-20S-Ergostane	$C_{28}H_{50}$	386	0.02		0.01	0.03	0.03	0.01	0.01		
14α,17β -20R-Ergostane	$C_{28}H_{50}$	386	0.02		0.02	0.00	0.03	0.01	0.01		
14α,17β -20S-Stigmastane	$C_{29}H_{52}$	400	0.03		0.03	0.06	0.06	0.02	0.02		
14β,17β -20R-Stigmastane	$C_{29}H_{52}$	400	0.05		0.05	0.12	0.10	0.03	0.02		
14β,17β -20S-Stigmastane	$C_{29}H_{52}$	400	0.05		0.04	0.12	0.07	0.03	0.03		
14α,17β -20R-Stigmastane	$C_{29}H_{52}$	400	0.05		0.05	0.07	0.06	0.03	0.00		
Total			0.50	0.00	0.77	0.66	0.67	0.24	0.23	0.44	0.29
% of total EOM			0.04	0.00	0.06	0.13	0.06	0.07	0.02	0.05	0.04
C_{27} S/(R + S)			0.29		0.64	0.33	0.32	0.61	0.27	0.41	0.17
C_{29} S/(R + S)			0.42		0.36	0.34	0.39	0.39	0.41	0.38	0.03
PAH											
Phenanthrene	$C_{14}H_{10}$	178	0.4	0.0	0.6	0.2	0.2	0.1	0.9		
Fluoranthene	$C_{16}H_{10}$	202	0.7	0.0	1.9	0.6	0.3	0.1	1.4		
Pyrene	$C_{16}H_{10}$	202	0.3	0.0	1.7	0.6	0.3	0.1	0.7		
Chrysene	$C_{18}H_{12}$	228	0.3	0.0	1.9	0.2	0.1	0.1	0.2		
1,3,5-Triphenylbenzene	$C_{24}H_{18}$	306	0.2	0	0.3	0.1	T	T	0.1		
Total			1.9	0.1	6.4	1.9	0.9	0.3	3.3	2.02	2.10
% of total EOM			0.12	0.03	0.48	0.35	0.08	0.09	0.34	0.21	0.17
Phenyl phosphates											
Triphenyl phosphate	$C_{18}H_{15}OP_4$	326	40.7	21.1	57.9	11.2	22.5	8.6	66.1		
Monotolyl diphenyl phosphate	$C_{19}H_{17}OP_4$	340	37.1	14.6	48.5	7.8	21.0	6.9	59.1		
Monotolyl diphenyl phosphate	$C_{19}H_{17}OP_4$	340	26.3	6.1	46.5	2.6	9.6	2.6	33.7		
Monophenyl ditolyl phosphate	$C_{20}H_{19}OP_4$	354	3.9	0.4	9.8	0.2	0.9	0.2	3.8		
Monophenyl ditolyl phosphate	$C_{20}H_{19}OP_4$	354	25.9	4.9	50.1	2.0	7.7	2.0	27.3		
Monophenyl ditolyl phosphate	$C_{20}H_{19}OP_4$	354	21.0	2.5	42.9	1.3	5.4	1.2	20.3		
Monophenyl ditolyl phosphate	$C_{20}H_{19}OP_4$	354	4.5	0.4	15.3	0.2	1.0	0.2	3.5		
Tri-m-cresyl phosphate	$C_{21}H_{21}OP_4$	368	2.6	0.4	6.1	0.1	0.4	0.1	1.6		
Tri-m-cresyl phosphate	$C_{21}H_{21}OP_4$	368	7.9	0.6	21.5	0.4	1.4	0.3	4.8		
Tri-m-cresyl phosphate	$C_{21}H_{21}OP_4$	368	7.9	0.7	25.6	0.3	1.6	0.3	5.4		
Tri-m-cresyl phosphate	$C_{21}H_{21}OP_4$	368	2.9	0.3	10.3	0.2	0.6	0.1	1.9		
Total			181	52	335	26	72	23	227	130.82	119.47
% of total EOM			12.9	18.4	26.1	5.2	6.6	6.5	23.9	14.23	8.68
Plasticizers											
Diethyl phthalate (DEP)	$C_{12}H_{14}O_4$	222	1.0	0.2	0.7	0.1	0.7	0.1	5.0		
Di-n-butyl phthalate (DBP)	$C_{16}H_{22}O_4$	278	29.7	18.8	36.1	10.1	28.2	13.2	44.5		
Diisobutyl phthalate (DIBP)	$C_{16}H_{22}O_4$	278	17.1	11.6	30.3	19.2	28.2	10.9	35.8		
Butyl benzyl phthalate (BBP)	$C_{19}H_{20}O_4$	312	0.8	2.8	1.6	0.9	0.0	0.6	3.5		
Dicyclohexyl phthalate (DCP)	$C_{20}H_{28}O_4$	330	0.0	0.0	0.6	0.5	0.8	0.0	0.0		
Diisoheptyl phthalate (DIHpP)	$C_{22}H_{34}O_4$	362	4.1	0.0	1.5	0.0	0.0	0.3	0.0		
Butyl decyl phthalate (BDP)	$C_{22}H_{34}O_4$	362	10.1	0.0	4.4	0.7	0.2	0.6	0.8		
Di(2-ethylhexyl) phthalate (DEHP)	$C_{24}H_{38}O_4$	390	11.6	21.1	20.3	13.2	21.6	9.1	19.1		
Di(n-octyl) phthalate (DNOP)	$C_{24}H_{38}O_4$	390	6.2	0.0	2.8	0.4	0.0	0.2	0.0		
Diisooctyl phthalate (DIOP)	$C_{24}H_{38}O_4$	390	5.1	0.0	2.1	0.3	0.0	0.0	0.0		
n-Octyl n-decyl phthalate (ODP)	$C_{24}H_{38}O_4$	390	15.2	0.5	8.9	0.8	0.5	0.7	0.8		
Diisononyl phthalate (DINP)	$C_{26}H_{42}O_4$	418	7.8	0.0	3.5	0.4	0.5	0.2	0.3		
Di(2-propyheptyl) phthalate (DPHP)	$C_{26}H_{42}O_4$	418	4.4	0.0	1.8	0.0	0.0	0.0	0.0		
Diisodecyl phthalate (DIDP)	$C_{28}H_{46}O_4$	446	13.8	0.3	6.9	0.7	0.0	0.5	0.6		
Diundecyl phthalate (DUP)	$C_{28}H_{46}O_4$	446	2.2	0.0	1.4	0.5	0.0	0.0	0.0		
Iso-*n*-undecyl phthalate	$C_{30}H_{50}O_4$	474	1.0	0.4	1.5	0.0	0.0	0.3	0.0		
Diisoundecyl phthalate (DIUP)	$C_{30}H_{50}O_4$	474	3.4	0.0	4.9	0.5	0.2	0.0	0.3		
Ditridecyl phthalate (DTDP)	$C_{30}H_{50}O_4$	474	3.8	0.0	0.9	0.1	0.0	0.1	0.1		
Diisotridecyl phthalate (DITP)	$C_{34}H_{58}O_4$	530	3.9	0.0	1.3	0.1	0.1	0.1	0.2		

(*continued on next page*)

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Table 1 (*continued*)

Compound	Composition	M.W.	Sampling date							Mean	SD
			23/6	3/7	6/7	20/7	17/8	8/9	14/9		
Total			141.0	55.7	131.7	48.5	81.1	36.9	111.0	86.56	41.84
% total EOM			10.1	19.7	10.3	9.5	7.5	10.7	11.7	11.34	3.90
n-Alkanols											
Tetradecanol	$C_{14}H_{30}O$	270	0.08		0.03	0.02	0.19	0.02	0.17		
Pentadecanol	$C_{15}H_{32}O$	256	0.00		0.00	0.00	0.06	0.00	0.01		
Hexadecanol	$C_{16}H_{34}O$	242	0.07		0.03	0.01	0.20	0.02	0.31		
Heptadecanol	$C_{17}H_{36}O$	256	0.00		0.00	0.00	0.03	0.00	0.02		
Octadecanol	$C_{18}H_{38}O$	270	0.12		0.06	0.05	0.78	0.06	0.38		
Nonadecanol	$C_{19}H_{40}O$	284	0.01		0.00	0.00	0.01	0.00	0.00		
Eicosanol	$C_{20}H_{42}O$	298	0.02		0.01	0.02	0.05	0.01	0.07		
Heneicosanol	$C_{21}H_{44}O$	308	0.01		0.00	0.01	0.02	0.00	0.01		
Docosanol	$C_{22}H_{46}O$	326	0.03		0.00	0.02	0.10	0.02	0.24		
Triacosanol	$C_{23}H_{48}O$	340	0.00		0.00	0.01	0.01	0.00	0.01		
Tetracosanol	$C_{24}H_{50}O$	354	0.10		0.01	0.03	0.08	0.02	0.05		
Pentacosanol	$C_{25}H_{52}O$	368	0.02		0.00	0.01	0.04	0.01	0.02		
Hexacosanol	$C_{26}H_{54}O$	382	0.20		0.03	0.08	0.33	0.05	0.15		
Heptacosanol	$C_{27}H_{56}O$	396	0.01		0.00	0.01	0.06	0.01	0.02		
Octacosanol	$C_{28}H_{58}O$	410	0.12		0.03	0.06	0.45	0.03	0.15		
Nonacosanol	$C_{29}H_{60}O$	424	0.01		0.00	0.01	0.08	0.01	0.02		
Triacontanol	$C_{30}H_{62}O$	438	0.09		0.04	0.05	0.77	0.03	0.18		
Hentriacontanol	$C_{31}H_{64}O$	452	0.00		0.00	0.00	0.01	0.00	0.00		
Dotriacontanol	$C_{32}H_{66}O$	466	0.00		0.00	0.00	0.05	0.00	0.00		
Tetratriacontanol	$C_{34}H_{70}O$	494	0.00		0.00	0.00	0.00	0.00	0.00		
Nonacosan-10-ol	$C_{29}H_{60}O$	424	0.00		0.00	0.00	0.00	0.00	0.00		
Total			0.89		0.25	0.38	3.33	0.29	1.80	1.16	1.21
% total EOM			0.06		0.02	0.07	0.31	0.08	0.19	0.12	0.11
CPI(e/o)			13.98		23.30	7.69	9.33	9.76	16.40	13.41	5.83
Methyl n-alkanoates											
Methyl nonanoate	$C_{10}H_{20}O_2$	172	0.1	0.0	0.3	0.0	0.4	0.9	0.4		
Methyl decanoate	$C_{11}H_{22}O_2$	186	0.1	0.2	0.4	0.3	0.4	0.9	0.8		
Methyl undecanoate	$C_{12}H_{24}O_2$	200	0.2	0.2	0.4	0.1	0.4	0.8	0.5		
Methyl dodecanoate	$C_{13}H_{26}O_2$	214	5.8	6.9	4.3	2.9	1.8	4.2	11.2		
Methyl tridecanoate	$C_{14}H_{28}O_2$	228	0.4	0.3	0.4	0.2	0.5	1.1	0.9		
Methyl tetradecanoate	$C_{15}H_{30}O_2$	242	2.2	2.9	1.3	2.8	1.4	3.3	3.4		
Methyl pentadecanoate	$C_{16}H_{32}O_2$	256	0.7	1.4	0.6	0.8	0.9	1.9	1.7		
Methyl hexadecanoate	$C_{17}H_{34}O_2$	270	9.6	15.1	7.2	9.4	9.3	21.2	12.9		
Methyl heptadecanoate	$C_{18}H_{36}O_2$	284	0.6	1.1	0.5	0.5	0.1	0.2	0.9		
Methyl octadecanoate	$C_{19}H_{36}O_2$	296	0.1	0.1	0.0	0.4	0.1	0.2	0.1		
Methyl octadecenoate	$C_{19}H_{38}O_2$	298	5.1	9.3	4.3	3.3	5.3	12.1	6.0		
Methyl nonadecanoate	$C_{20}H_{40}O_2$	312	0.4	0.4	0.4	0.1	0.2	0.6	0.3		
Methyl eicosanoate	$C_{21}H_{42}O_2$	326	0.7	1.0	0.4	0.6	0.6	1.3	0.6		
Methyl heneicosanoate	$C_{22}H_{44}O_2$	340	0.2	0.3	0.1	0.1	0.2	0.5	0.2		
Methyl docosanoate	$C_{23}H_{46}O_2$	354	0.5	1.0	0.2	0.5	0.5	1.2	0.4		
Methyl tricosanoate	$C_{24}H_{48}O_2$	368	0.2	0.4	0.1	0.1	0.3	0.6	0.2		
Methyl tetracosanoate	$C_{25}H_{50}O_2$	382	0.7	1.3	0.2	0.5	0.7	1.5	0.6		
Methyl pentacosanoate	$C_{26}H_{52}O_2$	396	0.1	0.4	0.1	0.1	0.2	0.5	0.2		
Methyl hexacosanoate	$C_{27}H_{54}O_2$	410	0.5	1.1	0.2	0.3	0.6	1.3	0.5		
Methyl heptacosanoate	$C_{28}H_{56}O_2$	424	0.1	0.3	0.0	0.1	0.2	0.5	0.2		
Methyl octacosanoate	$C_{29}H_{58}O_2$	438	0.4	1.5	0.2	0.4	0.7	1.5	0.5		
Methyl nonacosanoate	$C_{30}H_{60}O_2$	452	0.0	0.3	0.1	0.1	0.1	0.3	0.1		
Methyl triacontanoate	$C_{31}H_{62}O_2$	466	0.2	1.2	0.1	0.2	0.6	1.4	0.4		
Methyl hentriacontanoate	$C_{32}H_{64}O_2$	480	0.0	0.1	0.0	0.0	0.0	0.1	0.0		
Methyl dotriacontanoate	$C_{33}H_{66}O_2$	494	0.0	0.3	0.0	0.0	0.2	0.4	0.1		
Total			29.4	47.2	21.7	23.7	25.6	58.8	43.2	35.63	14.17
% total EOM			2.1	16.7	1.7	4.7	2.4	17.0	4.5	7.00	6.82
CPI(e/o)			1.85	1.38	1.84	2.22	1.27	1.27	2.17	1.71	0.41
Steroids											
Cholesterol	$C_{27}H_{46}O$	386	0.3		0.3	0.1	0.6	0.1	0.5		
Stigmasterol	$C_{28}H_{48}O$	412	0.2		0.1	0.0	0.1	0.0	0.1		
Sitosterol	$C_{29}H_{50}O$	414	0.5		0.2	0.0	0.4	0.0	0.2		

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Table 1 (*continued*)

Compound	Composition	M.W.	Sampling date							Mean	SD
			23/6	3/7	6/7	20/7	17/8	8/9	14/9		
Total			0.89	0.00	0.57	0.16	1.02	0.08	0.78	0.50	0.42
% total EOM			0.06	0.00	0.04	0.03	0.09	0.02	0.08	0.05	0.03
n-Alkan-2-ones											
Dodecan-2-one	$C_{12}H_{24}O$	184	0.2	0.0	0.0	0.1	0.2	0.1	0.0		
Tridecan-2-one	$C_{13}H_{26}O$	198	0.4	0.0	0.0	0.1	0.3	0.1	0.0		
Tetradecan-2-one	$C_{14}H_{28}O$	212	0.9	0.1	0.1	0.2	0.4	0.1	0.1		
Pentadecan-2-one	$C_{15}H_{30}O$	226	1.1	0.1	0.1	0.2	0.4	0.1	0.1		
Hexadecan-2-one	$C_{16}H_{32}O$	240	1.0	0.2	0.2	0.5	1.1	0.2	0.3		
Heptadecan-2-one	$C_{17}H_{34}O$	254	1.9	0.1	0.3	0.5	1.2	0.2	0.3		
Octadecan-2-one	$C_{18}H_{36}O$	268	1.0	0.1	0.2	0.4	1.1	0.1	0.2		
Nonadecan-2-one	$C_{19}H_{38}O$	282	1.1	0.2	0.5	0.8	1.9	0.2	0.4		
Eicosan-2-one	$C_{20}H_{40}O$	296	0.7	0.1	0.8	0.7	1.7	0.2	0.1		
Heneicosan-2-one	$C_{21}H_{42}O$	310	0.6	0.1	0.4	0.9	2.2	0.3	0.2		
Docosan-2-one	$C_{22}H_{44}O$	324	0.4	0.1	0.2	0.7	1.6	0.2	0.1		
Tricosan-2-one	$C_{23}H_{46}O$	338	0.4	0.1	0.1	0.5	1.2	0.1	0.1		
Tetracosan-2-one	$C_{24}H_{48}O$	352	0.3	0.0	0.1	0.4	0.9	0.1	0.1		
Pentacosan-2-one	$C_{25}H_{50}O$	366	0.3	0.0	0.1	0.3	0.7	0.1	0.0		
Hexacosan-2-one	$C_{26}H_{52}O$	380	0.3	0.0	0.1	0.3	0.6	0.1	0.0		
Heptacosan-2-one	$C_{27}H_{54}O$	394	0.2	0.0	0.1	0.2	0.6	0.1	0.0		
Octacosan-2-one	$C_{28}H_{56}O$	408	0.1	0.0	0.0	0.2	0.4	0.0	0.0		
Nonacosan-2-one	$C_{29}H_{58}O$	422	0.2	0.1	0.1	0.2	0.6	0.1	0.0		
Triacontan-2-one	$C_{30}H_{60}O$	436	0.1	0.0	0.0	0.1	0.3	0.0	0.0		
Hentriacontan-2-one	$C_{31}H_{62}O$	450	0.1	0.0	0.0	0.3	0.7	0.1	0.0		
Dotriacontan-2-one	$C_{32}H_{64}O$	464	0.0	0.0	0.0	0.0	0.1	0.0	0.0		
Tritriacontan-2-one	$C_{33}H_{66}O$	478	0.0	0.0	0.0	0.1	0.3	0.0	0.0		
Total			11.5	1.3	3.4	7.7	18.2	2.3	2.2	6.66	6.27
% total EOM			0.82	0.45	0.26	1.51	1.68	0.68	0.23	0.81	0.58
Total All Compounds			1403.1	282.7	1283.2	507.6	1083.9	346.0	951.0	836.79	456.49
UCM			867.7	110	721	301.9	729	187	508	489.25	296.04
% total EOM			61.8	38.8	56.2	59.5	67.3	54.0	53.4	55.86	8.96
UR			2.36	0.55	1.80	2.06	2.18	1.87	1.76	1.80	0.59

$a = \frac{\Sigma(C_{15}+C_{17}+\ldots+C_{33}+C_{35})}{\Sigma(C_{14}+C_{16}+\cdots+C_{32}+C_{34})}$, $b = \frac{\Sigma(C_{25}+C_{27}+\cdots+C_{33}+C_{35})}{\Sigma(C_{24}+C_{26}+\ldots+C_{32}+C_{34})}$ (Mazurek and Simoneit, 1984).
0.0 or blank = not detected.

(Mazurek and Simoneit, 1984; Rogge et al., 1993a; Simoneit and Mazurek, 1982), and were calculated for the entire C_{15}–C_{35} range and for C_{24}–C_{35}. The CPI values varied from 0.9 to 1.7 (mean = 1.4 ± 0.3) for the full range and from 0.8 to 1.9 (mean = 1.4 ± 0.4) for the C_{24}–C_{35} range (Table 1). Generally, a CPI value of unity indicates a petroleum source. The distributions of *n*-alkanes and low CPI values in these samples support major anthropogenic sources. These CPI values are similar to values for aerosols from other urban regions (e.g. Bi et al., 2008; Didyk et al., 2000; Rogge et al., 1993b; Simoneit et al., 1988; Zhou et al., 2008, 2009). The sources of the *n*-alkanes can be from incomplete combustion of fossil fuels and waxes from natural vegetation. The natural vegetation waxes consist of the longer chain ($>C_{20}$) *n*-alkanes, *n*-alkanoic acids and *n*-alkanols (Simoneit and Mazurek, 1982). The concentrations of the plant wax alkanes were calculated by subtraction of the smooth alkane envelope from the total alkanes (Simoneit et al., 1991a), and were found to range from 4 to 30 ng m^{-3} (mean = 14 ± 10 ng m^{-3}). The contribution of *n*-alkanes attributed to vegetation wax is major in these samples as shown by the significant positive correlation (R^2 = 0.70) between *n*-alkanes from waxes and total *n*-alkanes (Fig. 4a). The CPI values obviously increase with the increase of *n*-alkane wax inputs from vegetation, as shown in Fig. 4b. The significant positive correlation (R^2 = 0.80) between the CPI and *n*-alkane wax validates the power of the CPI parameter as an indicator for distinguishing between natural and anthropogenic sources.

The geochemical alteration of 3,7,11,15-tetramethyl-2-hexadecen-1-ol (phytol), produces 2,6,10,14-tetramethylpentadecane (pristane, Pr) and 2,6,10,14-tetramethylhexadecane (phytane, Ph) (Bendoraitis et al., 1962; Maxwell et al., 1971; Ikan et al., 1975; Didyk et al., 1978). Pr and Ph are found in lubricating oil, crude oil, diesel fuel and in both diesel and auto engine exhaust (Simoneit, 1984, 1985). The concentrations of these branched hydrocarbons in the aerosol PM samples are given in Table 1. The occurrence of pristane, phytane and UCM generally confirms the presence of residues from fossil fuel and petroleum products (Simoneit, 1984, 1985). The ratio of pristane-to-phytane (Pr/Pr) ranged from 0.11 to 1.20 with a mean of 0.62 ± 0.53, indicating that the major source is petroleum products.

The UCM concentrations varied from 110 to 1408 ng m^{-3} (mean = 489 ± 296 ng m^{-3}) (Table 1). The presence of an

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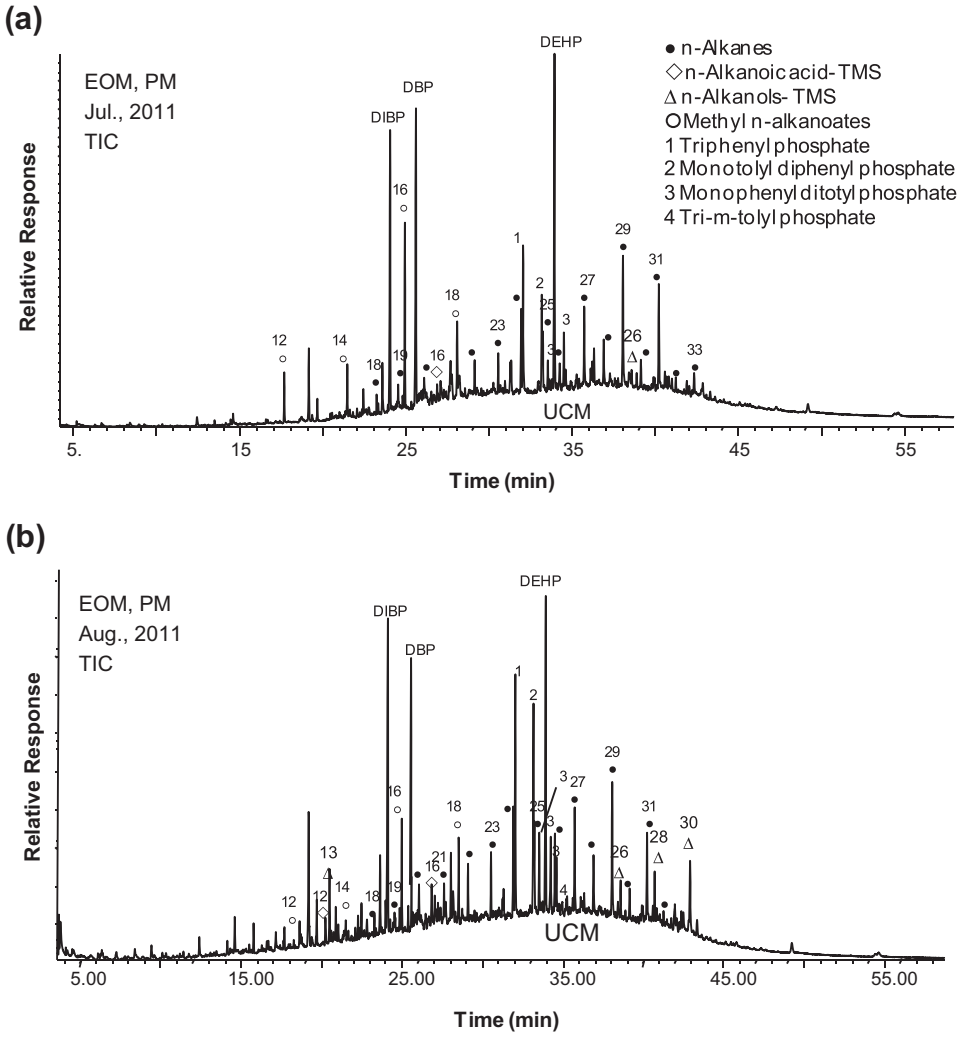

Figure 2 GC–MS total ion current (TIC) traces of typical total extracts from aerosol PM samples showing the major organic components (numbers refer to the carbon chain length, DIBP = di-isobutylphthalate, DBP = dibutylphthalate, DEHP = diethylhexyl phthalate, and UCM = unresolved complex mixture): (a) July 2011, and (b) August 2011.

UCM in aerosol PM generally indicates inputs from lubricants in combustion engine emissions, i.e. traffic (Simoneit, 1984, 1985). These concentration values of UCM and *n*-alkanes were similar to those for other large cities, e.g. Santiago (Chile), Los Angeles (CA), Guangzhou (China), Beijing (China), and Ibadan (Nigeria) (Bi et al., 2008; Didyk et al., 2000; Rogge et al., 1993b; Simoneit et al., 1988, 1991b; Zhou et al., 2008, 2009). The ratio of unresolved (i.e. UCM) to resolved compounds, *U*:*R*, is another parameter for gauging contamination by fossil fuel residues (Mazurek and Simoneit, 1984). The *U*:*R* for these samples varied from 0.55 to 2.06 (mean = 1.80 ± 0.59, Table 1), also falling in the range of that observed for the other cities mentioned above.

The hopane and sterane hydrocarbons, which are other characteristic petroleum indicators (Simoneit, 1984, 1985; Peters and Moldowan, 1993), were evident in these samples (Table 1). The concentrations of the hopanes ranged from 0.4 to 18.7 ng m^{-3} (mean = 11.2 ± 8.4 ng m^{-3}), whereas the steranes had a lower range from 0.0 to 0.8 ng m^{-3} (mean = 0.4 ± 0.3 ng m^{-3}). Therefore, the presence of hopanes and steranes, which are usually resistant to degradation in the environment, further confirms contamination from fossil fuel residues (e.g. Simoneit, 1984; Simoneit et al., 1991b; Medeiros et al., 2005). The major compounds of the hopanes had the thermodynamically more stable 17α(H),21β(H) configuration, maximizing at the C_{30} homolog, and minor 17β(H),21α(H)-hopanes. Their distribution shows that the 17α(H),21β(H)-hopane series, ranging from C_{27} to C_{35}, is the predominant series with the homologs $> C_{31}$ present as the typically mature C-22 *R*/*S* pairs (Simoneit, 1984; Simoneit et al., 1990). Distributions showing higher concentrations of the 22*S* hopanes relative to the corresponding 22*R* pairs are commonly from gasoline and diesel engine exhaust (Simoneit, 1984, 1985).

The $S/(S + R)$ ratios for C_{31} and C_{32} extended hopanes ranged from 0.56 to 0.60 (mean = 0.58 ± 0.01) and from 0.59 to 0.64 (mean 0.61 ± 0.02), respectively. These values are comparable to those for petroleum hydrocarbons and mature crude oil (Kvenvolden et al., 1990; Rushdi and Simoneit, 2002a,b), and therefore indicate the origin of the hopanes to be from petroleum.

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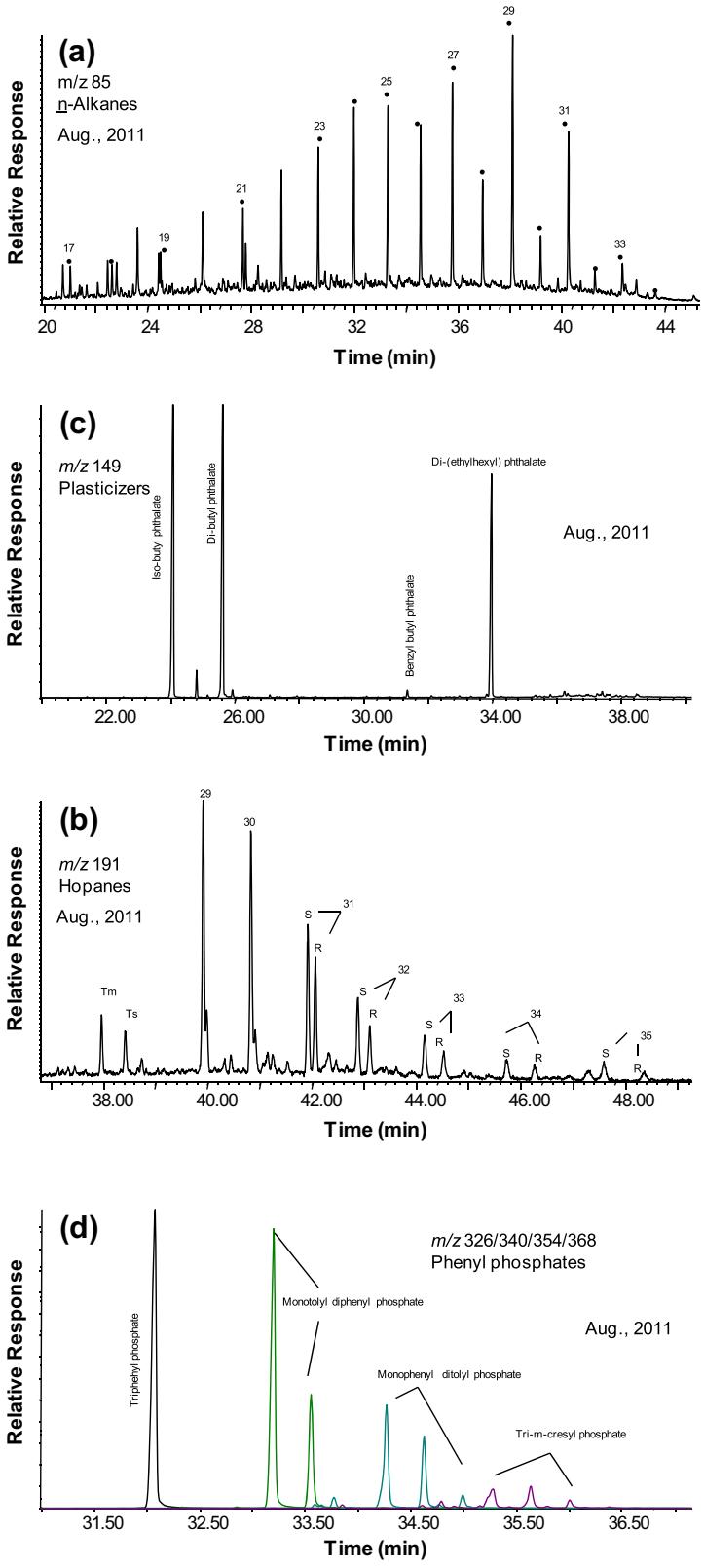

Figure 3 Examples of typical GC–MS key ion plots (August 2011 sample): (a) *n*-alkanes (*m*/*z* 85), (b) hopanes (*m*/*z* 191), (c) plasticizers (*m*/*z* 149), and (d) phenyl phosphates (*m*/*z* 326/340/354/368).

Steranes are not common in gasoline or diesel fuels and are introduced into the environment directly from petroleum or from lubricants in emissions of vehicular engines (Abas and Simoneit, 1996). Municipal wastewaters and refinery activities are also possible sources for a significant amount of petroleum product inputs to the atmosphere. Thus, steranes are also

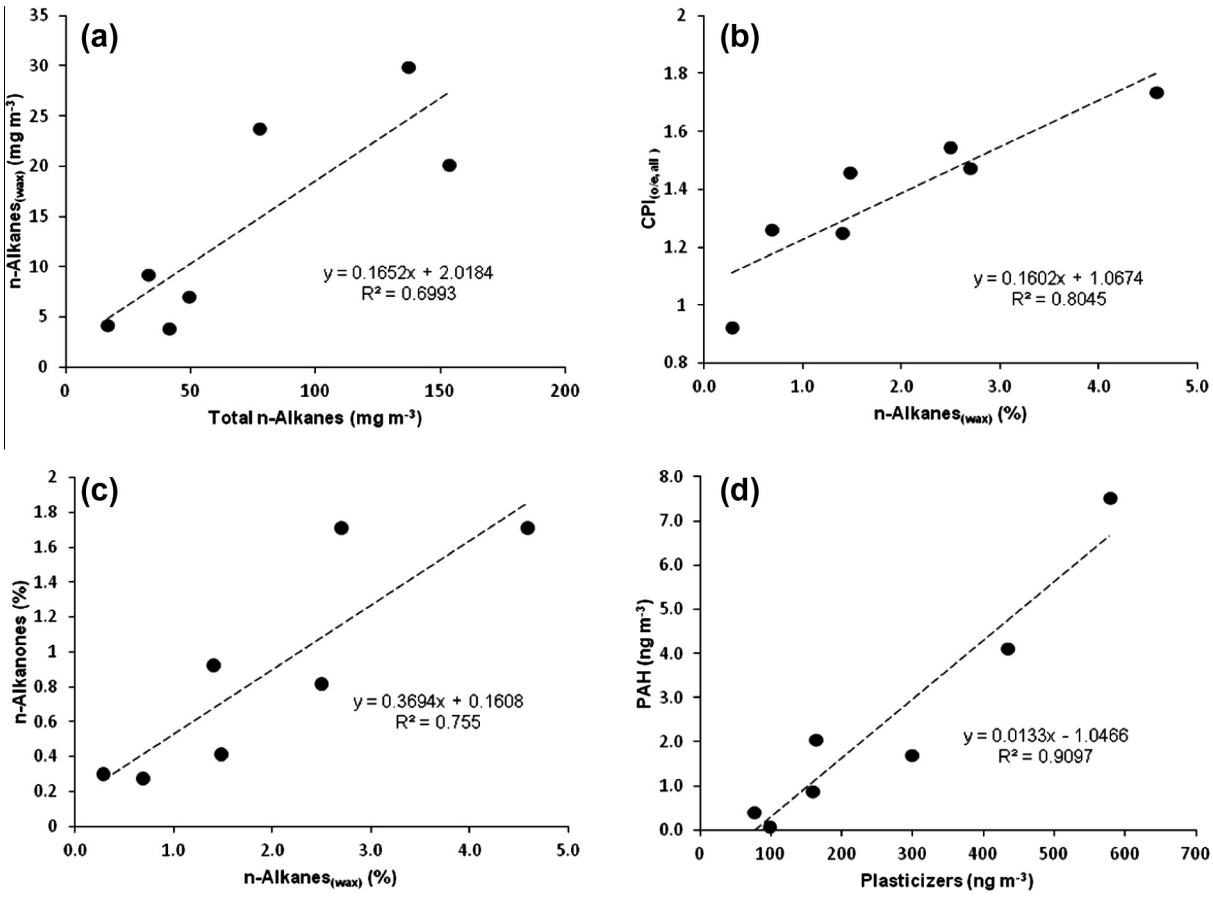

Figure 4 Correlation plots showing the relationships of: (a) the concentration of wax *n*-alkanes versus total *n*-alkanes, (b) CPI values versus fractions of wax *n*-alkanes, (c) fractions of *n*-alkanones versus wax *n*-alkanes, and (d) concentrations of total PAHs versus plasticizers.

useful indicators for oil-product pollution in urban areas (e.g. Albaiges and Cuberes, 1980; Aboul-Kassim and Simoneit, 1996). The steranes in these PM samples ranged from C_{27} to C_{29} with mainly the 5α,14α,17β and minor 5α,14α,17α configurations, both occurring as 20S and 20R epimers. The sterane epimerization ratio at C-20, $S/(S + R)$ for these samples range from 0.29 to 0.61 (mean = 0.41 ± 0.17) for C_{27} and from 0.34 to 0.42 (mean = 0.38 ± 0.03) for C_{29}. These values indicate that the main sources of these biomarkers are from petroleum sources.

4.2. Polycyclic aromatic hydrocarbons (PAHs)

PAHs are produced from incomplete combustion of any organic material. They are considered as serious atmospheric pollutants, because some PAHs are well known as human carcinogens (IARC, 1991). The major sources of PAH emissions are natural from forest fires (Simoneit, 2002; Vila-Escalé et al., 2007), and anthropogenic from industries, vehicles and burning of fossil and biomass fuels (e.g. Rogge et al., 1993a; Simoneit et al., 2005, 2007; Yang et al., 2002). The concentrations and the dominant individual PAHs measured in this study are given in Table 1. Only phenanthrene (Phe), fluoranthene (Fl), pyrene (Py) and chrysene (Chry) were detected in these samples. It should be mentioned that 1,3,5-triphenylbenzene, a PAH from burning of plastics (Simoneit et al., 2005), is discussed below. The concentrations of PAHs were relatively low and ranged from 0.1 to 6.1 ng m^{-3} (mean = 2.0 ± 2.1 ng m^{-3}). The low concentration of PAHs may be due to higher temperatures and possibly higher wind speeds or enhanced photo-degradation of PAHs, which lead to increased dispersion and decrease in the total PAHs. Additionally, most of the PM samples were dominated by 3 and 4 ring PAHs typical of freshly emitted smoke (Medeiros and Simoneit, 2008; Oros and Simoneit, 2001a,b). Burning of plastics and garbage waste may be the main source of these PAHs in the PM samples.

4.3. Polar compounds

The fatty acids (*n*-alkanoic acids) of flora and fauna usually range from C_{12} to C_{32} with even carbon chain length homologs. They are commonly unsaturated in flora where the C_{18} mono-, di- and tri-unsaturated compounds are generally the major fatty acids. Because fatty alcohols (*n*-alkanols) in plants are biosynthesized from fatty acids by enzymatic reduction, they have similar carbon chain lengths with predominantly even number chains (Lehninger, 1970; More, 1993). The presence of *n*-alkanoic acids ($< C_{20}$) in the environment, with an even carbon number predominance and C_{max} at 16 indicates multiple sources from mainly microbial and lesser vascular plant detritus (Simoneit, 1977, 1989). The presence of unsaturated fatty acids $< C_{20}$ reflects fresh OM sources not subjected to oxidation and biodegradation in the environment.

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The total *n*-alkanoic acids (analyzed as methyl *n*-alkanoates) were significant components in these samples (7.0 ± 6.8% of EOM), with concentrations ranging from 21.7 to 58.8 ng m^{-3} (mean 35.6 ± 14.2 ng m^{-3}) (Table 1). These values are similar to other urban areas (e.g. Abas and Simoneit, 1996; Didyk et al., 2000; Rogge et al., 1993b; Schauer et al., 1996). The *n*-alkanoic acids of these samples ranged mainly from C_9 to C_{32} with a maximum concentration at C_{17} (C_{max} at 16 as acid, Fig. 2c). The concentrations of the unsaturated acids (comprised mainly of $C_{18:1}$) were low ranging from nil to 0.4 ng m^{-3}, indicating a rapid removal of these alkenoic acids by photo-oxidation or probably the occurrence of aged aerosol particles.

The *n*-alkanols were detected at low amounts in these samples. They ranged from nil to 3.3 ng m^{-3} with a mean value of 1.16 ± 1.21 ng m^{-3}. The maximum concentrations were observed at C_{16}, C_{18}, C_{26} or C_{30}. This *n*-alkanol distribution indicates an input of vascular plant wax from semitropical to tropical environments (Simoneit, 1977, 1989; Rushdi et al., 2006).

Steroids are derived from biogenic sources, occur in all ecosystems, and can be utilized to identify the origin and fate of OM in the environment (Volkman et al., 1981; Rushdi et al., 2006). They comprise a variety of compounds and generally range from C_{26} to C_{30} (Moreau et al., 2002). Cholesterol (C_{27}) is a major compound in faunal lipids, plankton and some terrestrial plants. Steroids in higher plants are known as phytosterols and they range from C_{28} to C_{30} with one or two carbon–carbon bonds, typically one in the sterol nucleus and a second in alkyl side chain (Brassell et al., 1983). Steroids were present at low concentrations in these samples and included cholesterol, stigmasterol and sitosterol, with concentrations ranging from 0.0 to 1.02 ng m^{-3} (mean = 0.5 ± 0.4 ng m^{-3}) (Table 1). The dominance of stigmasterol and sitosterol in the samples is interpreted as an origin from terrigenous higher plants (Barbier et al., 1981; Simoneit et al., 1983; Moreau et al., 2002).

The *n*-alkan-2-ones were present in these aerosol PM samples, ranging from C_{12} to C_{33}, with concentrations of 1.3–18.2 ng m^{-3} (mean = 6.7 ± 6.3 ng m^{-3}) (Table 1). The *n*-alkanones are indicators for terrigenous inputs, possibly from resuspended soil dust, because they are found in terrestrial soils and vegetative detritus, and may form by microbial processes from the *n*-alkanes of plant wax (e.g. Morrison and Bick, 1966; Oros et al., 2002). The significant positive correlation (R^2 = 0.76) between total concentrations of *n*-alkanones and wax *n*-alkanes (Fig. 4c) indicates that they are likely from the same source.

4.4. Synthetic organic compounds

Chemical industries produce numerous and varied amounts of synthetic organic compounds. These chemicals are used for different purposes and may have health impacts if they are unwisely disposed into the environment. Plasticizers, silicones, and other synthetic organic compounds from fugitive sources can be part of the components of atmospheric PM in urban areas. Direct out-gassing of vinyl chloride and other volatile compounds, and burning of plastics (Simoneit et al., 2005) are the major sources of plasticizers in the atmosphere.

The total plasticizers in these PM aerosol samples varied from 75 to 338 ng m^{-3} (mean = 217 ± 156 ng m^{-3}) (Table 1). Thus, the aerosols of the city had 14.1–35.6% (mean = 25.5 ± 10.8%) of EOM attributable to synthetic compounds. Phenyl phosphates were the dominant plasticizers in these samples with concentrations ranging from 23 to 227 ng m^{-3} (mean = 130.8 ± 119.5 ng m^{-3}) (Table 1). Phthalates were also major synthetic organic compounds in these samples, ranging from 36.9 to 141.0 ng m^{-3} (mean = 86.9 ± 41.8 ng m^{-3}) (Table 1). The dominant compounds found in most of the samples were triphenyl phosphate (TPP), di-*n*-butylphthalate (DBP), di-isobutylphthalate (DIBP) and di(2-ethylhexyl)phthalate (DEHP) (Table 1). Low levels of 1,3,5-triphenylbenzene were present in most samples (trace-0.4 ng m^{-3}, Table 1). This compound is a specific tracer from burning of plastics (Simoneit et al., 2005), and its occurrence in these samples confirmed that such materials were burned in the vicinity of Dhahran city. The positive correlation (R^2 = 0.91) between the concentrations of total PAHs and plasticizers (Fig. 4d) confirmed that they were from the same source and likely in part a result of burning practices. The levels of plasticizers in these aerosols indicate that the city is obviously polluted by significant direct emissions and from burning of plastics and other wastes.

Recent results show that phthalate (e.g. DBP, DEHP and DINP) exposure affects human reproductive development (Swan, 2008; Swan et al., 2005; Howdeshell et al., 2008) and can result in estrogenic consequences (Jobling et al., 1995). Therefore, long term exposure to these plasticizers may have a significant impact on the health of the local population.

4.5. Source apportionment

The mean concentrations of the major organic compound groups in the aerosol PM of the Dhahran city were found to be lower relative to other large cities (Abas and Simoneit, 1996; Didyk et al., 2000; Giri et al., 2013; Rogge et al.,

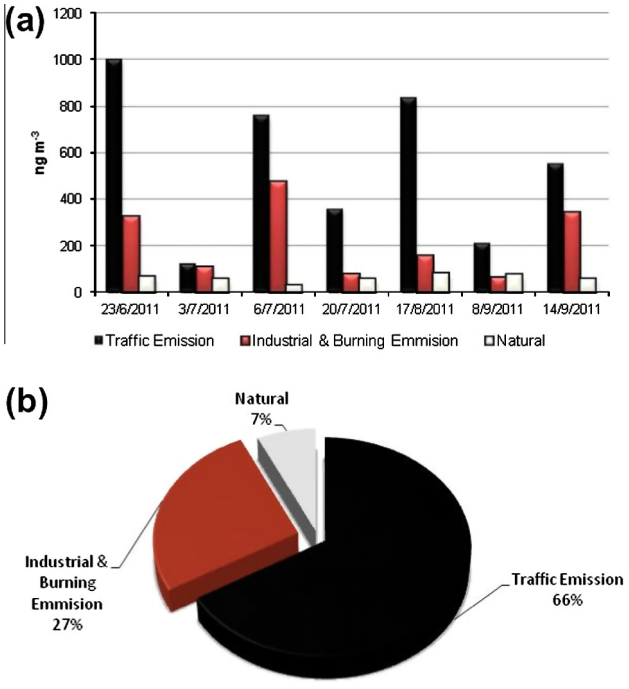

Figure 5 Plots showing the percent fractions of traffic emissions, industrial and burning emissions, and natural background for (a) different sampling dates and (b) the average of all samples.

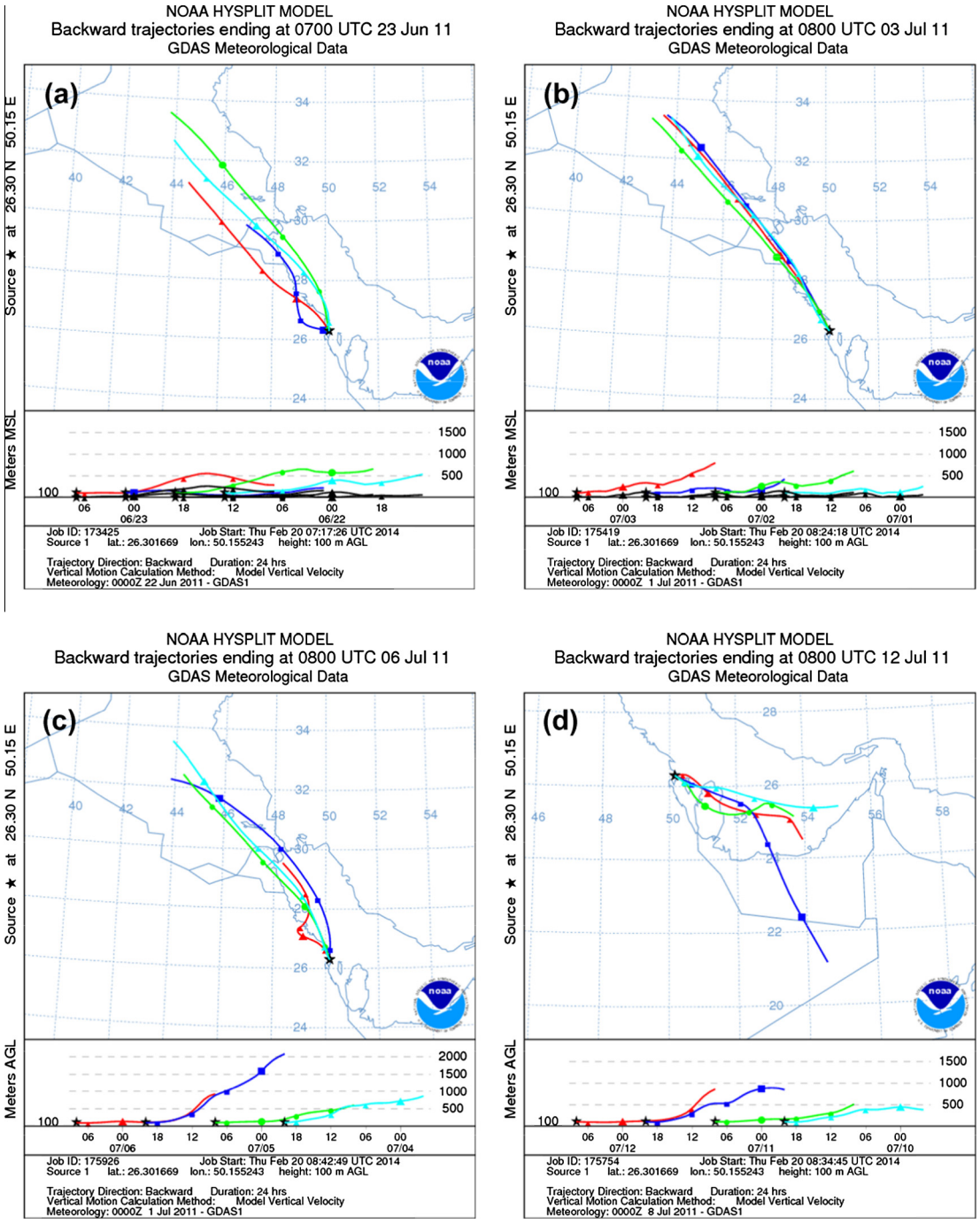

Figure 6 Backward trajectories in the year 2011 using NOAA Hysplit Model for (a) June 23, (b) July 3, (c) July 6, (d) July 12, (e) July 20, (f) August 17, (g) September 8, and (h) September 14.

1993b). The mean UCM value (489 ± 296 ng m^{-3}) is considerably lower than those of other cities, indicating lower emissions of lubricating oil from vehicular traffic in the city of Dhahran (Simoneit, 1985). The plasticizer levels are significant in Dhahran, but still lower than other large industrial cities (e.g. Giri et al., 2013). The *n*-alkanoic acids and *n*-alkanols in the PM are much lower than for other cities due to the low vegetation growth in this region. The organic compound group concentrations for Dhahran aerosol PM have been summed for the major emission source categories as follows:

(1) Emissions from petroleum product utilization (i.e. traffic and industrial engines) are comprised of UCM, biomarkers (hopanes and steranes), and total *n*-alkanes less the plant wax *n*-alkanes.
(2) Burning and industrial emissions from plastics and municipal waste burning (Didyk et al., 2000; Simoneit et al., 2005), and direct volatilization of plasticizers. It should also be noted that the low amounts of PAHs, with triphenylbenzene, are likely derived from burning of plastics.

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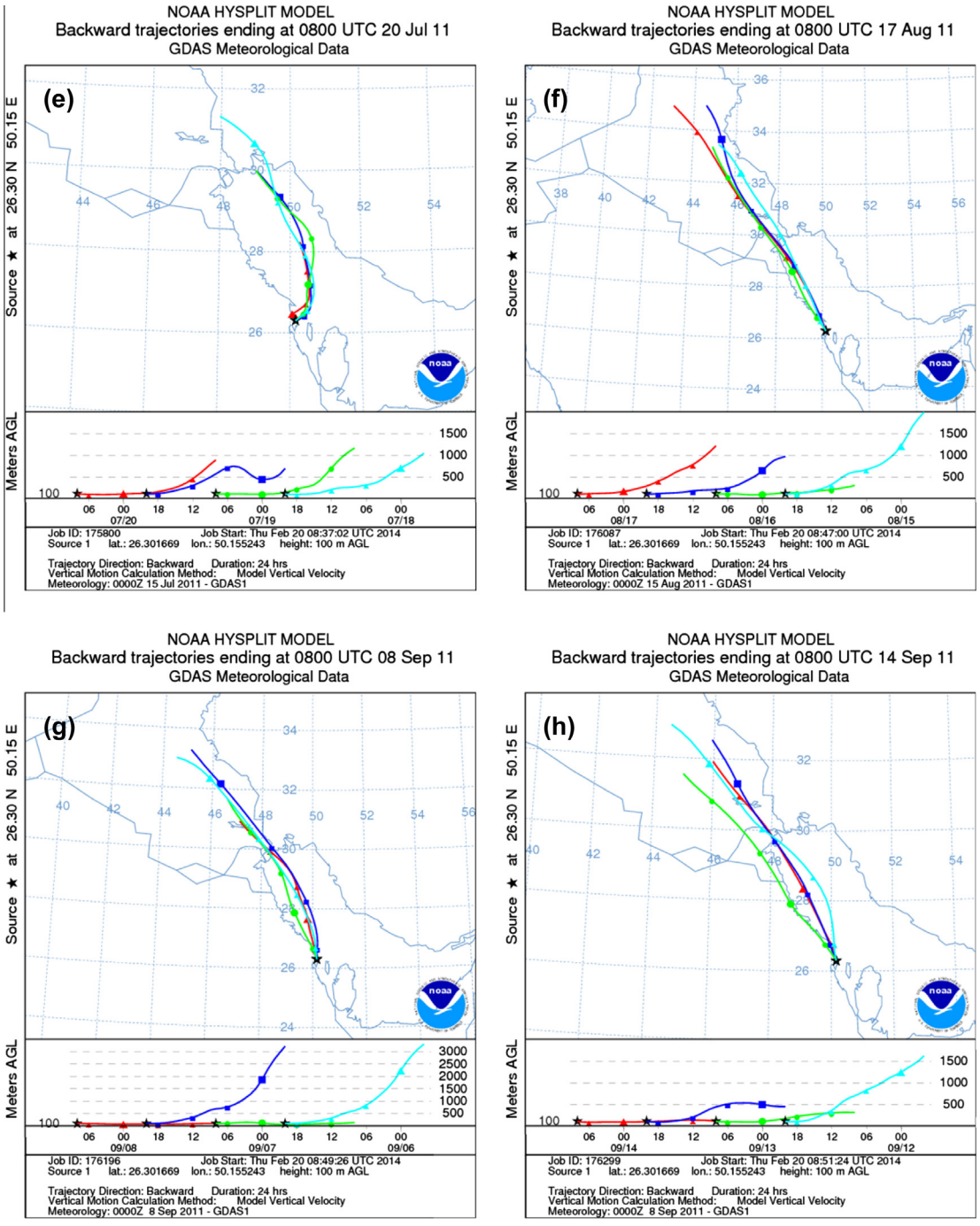

Fig. 6 (*continued*)

(3) Natural inputs of higher plant waxes from surrounding vegetation. They are the sum of long chain ($>C_{20}$) *n*-alkanes, *n*-alkanones, and methyl *n*-alkanoates (Simoneit, 1989).

The source emission categories are quite diverse as shown in Fig. 5, where the major contributions for all samples are from traffic emissions, and plastic and waste burning. Internal combustion engine emissions are a dominant source and comprise 43–78% (mean = 64 ± 12%) of the total extractable organic matter (EOM). Emissions from plastic and waste burning are also a major source, ranging from 14% to 38% (mean = 26 ± 11%) of the total EOM. The natural background is relatively low for all samples ranging from 2% to 21% (mean = 10 ± 7%) of the total EOM. Generally, anthropogenic emissions dominate the total EOM of aerosol PM in Dhahran city, where concern and additional work by local authorities are warranted.

The backward trajectory analyses show that the air masses generally approached the city of Dhahran from the northwest in Iraq during the sampling periods as illustrated in Fig. 6. On June 23rd, July 3rd, July 6th, August 17th, and part of the time

on September 14th the air masses were mainly inland (Fig. 6a–c,f,h) leading to relatively high concentrations of organic tracers in the aerosol PM and supporting a superimposed external PM transport contribution to the ambient Dhahran PM. On July 20th and September 8th the air masses were offshore (Fig. 6, e.g) with low concentrations of organic compounds in the aerosol PM. This indicates that the origins and directions of the air masses drifting into the sampling region may be significant contributors to the quality and quantity of organic tracers in aerosol PM. Simultaneous sampling needs to be conducted upwind of the city and in Dhahran to correct for the external input contribution.

5. Conclusions

The organic compounds, including plasticizers, *n*-alkanes, plant wax alkanes, PAHs, *n*-alkanoic acids, *n*-alkanones, and steroids, in aerosol PM were investigated for Dhahran, Saudi Arabia. The compounds from plastics were significant in all samples due to their wide production, use and disposal in this urban/industrial region. Combustion emissions from mainly fossil fuel product use and extensive waste burning were the dominant sources for the organic compounds in this aerosol PM. Natural (background) organic compounds were relatively low. Further studies are needed to investigate the externally sourced drift-in of PM, and the occurrence, composition and toxicity of the components in the various size fractions of aerosol PM (i.e. $PM_{2.5}$ and PM_{10}) in the main cities of Saudi Arabia.

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